Observation of new doubly-hollow lithium states produced by triple excitation of lithium atoms in the ground state

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INTRODUCTION

A hollow atomic state is an excited state in which the first inner-shell (the 1s-shell) is empty. Correlations effects can produce simultaneous triple excitation of all three electrons in lithium atoms and create hollow lithium states of the type (nl, n'l', n''l'') with n, n' and n'' at least equal to 2. After the first observation of the lowest-energy triply excited state[1], several experiments involving the use of photoion[2,3] and photoelectron[4-6] spectrometries have provided the energy of a number of such (nl, n'l', n''l'') states and partial cross sections for photoionization of lithium atoms into various continua of the Li^+ ion). Theoretical calculations using the R-matrix approximation have provided results for the partial photoionization cross sections[5] that are generally in good agreement with the experimental data, whereas the saddle-point technique[7] has given very accurate calculations of the energies of many hollow lithium states.

The demands on theoretical and experimental techniques increase at higher photon energies, for which triply excited states with all three electrons having principal quantum numbers equal to 3 or above can be created. The research of such excited states is particularly challenging as cross sections for these states decrease fast and configuration excitation is especially strong in the theoretical calculation. We have proposed[8] the name of doubly-hollow state for triply excited states in which the two first inner-shells, the K- and L-shells, are empty. In previous experiments,[8] the lowest-energy doubly-hollow ($3s^23p$ ^2P in the one-electron notation) lithium state was measured using electron spectroscopy together with the synchrotron radiation emitted by the Advanced Light Source in Berkeley. The energy measured for this state was 175.17(5) eV. Simultaneously, a photoabsorption experiment using the Photon Factory in Tsukuba provided results in good agreement with our observation.[9] In the work done at ALS during the year 2000 and briefly presented here, we have succeeded in measuring new doubly-hollow states in lithium of the type (3l, 3l', 3l'').

EXPERIMENT

The experiments were carried out at the AMO undulator beam line 10.0.1 of the ALS. The spherical grating monochromator was used with the 2100 lines/mm grating to provide photons with energies between 175 and 180 eV, and with bandwidths in the range of 80 to 100 meV. The electron spectra were meaured using the Scienta-200 hemispherical analyzer designed for gas phase energy-and angle-resolved studies. The analyzer was operated at a constant pass energy of 40 eV with an electron energy resolution of 40 meV. An effusive beam of lithium atoms was produced by a resistively heated oven placed at right angle to the direction of the photon beam. Liquid nitrogen cooled traps prevented the lithium vapor to deposit on the electrodes within the zoom lens of the electron spectrometer, The spectra presented here are not corrected for the transmission of the electron analyzer.

EXPERIMENTAL PROCEDURE

At excitation energies higher than all doubly-excited (2l, n'l') states of Li^+, triply excited atomic states can decay to either singly-excited (1s, nl) or doubly-excited (2l, 2l') and (2l, 3l') states of Li^+. Decay path into (1s, nl) Li^+ ionic states causes interferences with the direct photoionization route as reported earlier.[8] Decay to doubly-excited (2l, n'l') Li^+ states produces emisssion of a low-energy electron. These Li^+ states subsequently decay to the ground state of Li^{2+} ion, with emission of a high-energy Auger electron.[10] In our experiments, we chose to measure the high-energy Auger electron spectrum resulting from the decay of the (2l, 3l') Li^+ states, since theoretical R-matrix calculations predicted these channels to be favorably populated in the decay of the doubly-hollow states.

RESULTS

We show in Figure 1 an example of the Auger spectra emitted in the decay of the (2l, 3l') excited state of Li^+. The kinetic energy of these Auger electrons is between 81 and 85 eV, corresponding to the difference between the energy of the doubly-excited Li^+ state and the energy of the ground state of Li^{2+}. Peaks are noted from 1 to 9, according to increasing kinetic energies. The assignment and the measured energy of these peaks has been previously published.[10] This Auger spectrum has been measured measured at 177 eV photon energy, i. e., at an excitation energy where only direct photoionization can populate the doubly-excited states of Li^+. Knowing from R-matrix calculations[11] the predicted energy of the doubly-hollow states, we varied continuously the photon energy from 177 to 179 eV, measuring simultaneously the (2l, 3l') Auger spectrum in using the CIS mode of the Scienta. In this way we obtained the photon energy dependence of the intensity of every Auger electron peak visible in Figure 1. As an example, we show in Figure 2 the result of the measurements for peak number 6 which is due to the decay of the 2p3p ^1D state, also noted (0, 1, 0) in the hyperspherical coordinates introduced by Lin.[12] The relative intensity of this peak has been normalized to the intensity of peak number 1 (2s3s ^3S or (1, 0, -) state)). In this branching ratio, we see distinctly two broad axima whose energies were measured to be 177.75 (10) and 178.18 (12) eV. These structures are due to interferences between the direct photoionization route and the decay of the doubly-exctited state resonantly populated by the decay of doubly-hollow states, but they are dominated by the resonant process. We measured the same behavior for peaks 4, 7 and 8, while in other continuum channels (peaks 5 and 9), only one broad maximum was observed), reflecting the differential decay of the various doubly-excited states in the different continua. In this energy range, the saddle-point complex rotation method used by Chung[13] predicted the existence of one resonance at 176.88 eV, while R-matrix calculations by Lan[11] calculated this resonance to be at

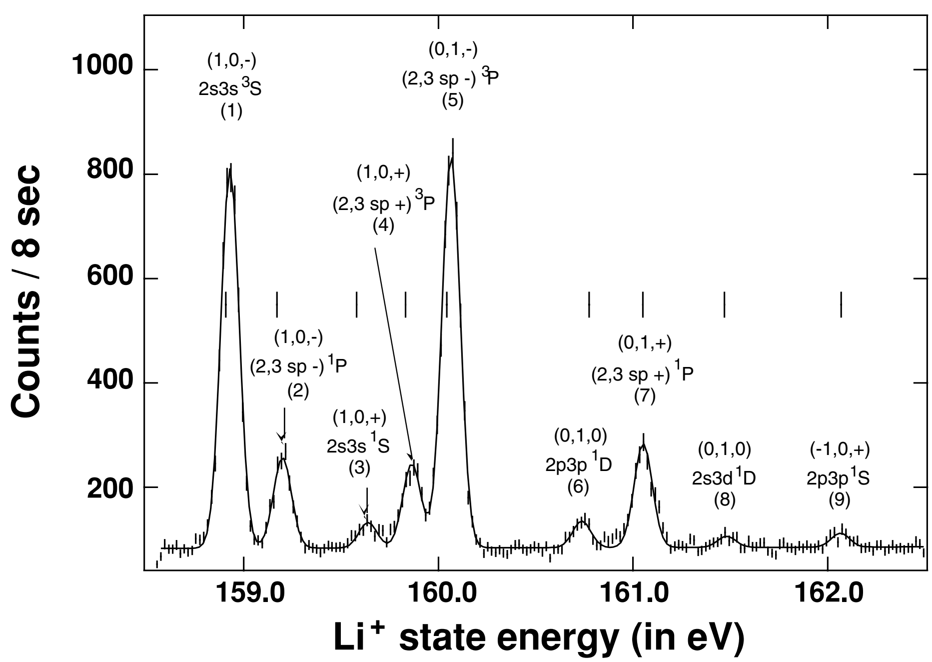

Fig. 1. Auger spectrum from (2l, 3l') Li^+ states.

177.73 eV (3s3p3d ^{2}P triply excited state) and predicted an additional one at 178.09 eV (3s3p4s ^{2}P state). In order to determine the number and the energy of the doubly-hollow states in this energy range, we need to know the relative cross section for photoionization into every channel. This remains to be done, however, since, in the experiments presented here, the fast variation of the density of lithium vapor in the interaction region prevented us to determine with reasonable accuracy the partial photoionization cross sections.

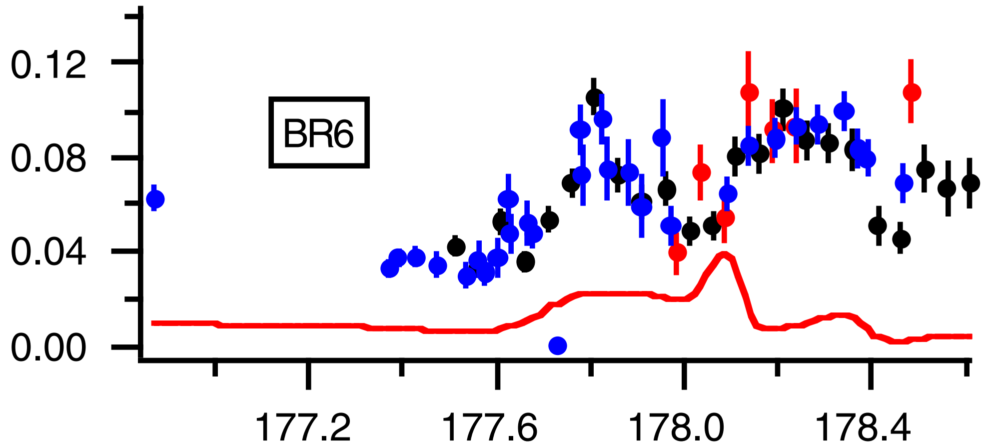

Fig. 2. Energy dependence of the branching ratio between the intensity of peak number 6 in figure 1 (from 2p3p ^{1}D Li$^+$ state) amd peak number 1 (from 2s3s ^{3}S Li$^+$ state).

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